

# Spin relaxation in a two-electron quantum dot

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We discuss the rate of relaxation of the total spin in the two-electron droplet in the vicinity of the magnetic field driven singlet-triplet transition. The total spin relaxation is attributed to spin-orbit and electron-phonon interactions. The relaxation process is found to depend on the spin of ground and excited states. This asymmetry is used to explain puzzles in recent high source-drain transport experiments.

PACS numbers: 73.20.Dx, 71.50.+t. *Keywords:* Spin relaxation; Quantum dots; Singlet-triplet transition; High magnetic field.

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The electronic structure of artificial atoms in quantum dots (QD's) [1] can be directly studied in transport measurements. Such voltage-tunable experiments in a varying magnetic field (e.g., see Refs. [1–7] and the works cited therein) enable to change the number of electrons confined in a QD as well as the mutual arrangement of electronic energy levels. The arrangement of levels depends on the total spin of electronic configurations [3,4]. The application of the magnetic field induces transitions in the ground state between states characterised by different total spin. In this paper we focus on a role played by total spin in a simplest system, a two-electron droplet. At low magnetic fields the ground state is a singlet with total spin  $S = 0$  while at higher magnetic fields the ground state is a triplet with total spin  $S = 1$ , in analogy to the parahelium-orthohelium transition, already studied theoretically in Refs. [4,8]. Both singlet and triplet states and the singlet-triplet (ST) transition have been observed experimentally in both vertical [2,6] and lateral QD's [5,7]. However, while at least one of the triplet excited state is observed for magnetic fields below the ST transition, the singlet excited state is not seen past the ST transition, resulting in asymmetric (in  $B$ ) high source and drain transport spectra. In the present work we present model calculations of total spin relaxation due to the mixture of spin-orbit (SO) and electron-phonon interaction which helps to explain the unusual behaviour of the levels associated with different total spin seen in transport experiments.

We start with the SO interaction Hamiltonian for a two-dimensional (2D) electron in a quantum well written as [9–13]:

$$H_{SO} = \alpha (\mathbf{k} \times \hat{\boldsymbol{\sigma}})_z - \beta (\bar{\mathbf{k}} \cdot \hat{\boldsymbol{\sigma}}) , \quad (1)$$

where the layer plane is determined by the principal axes  $(x, y)$  of the crystal. This expression is a combination of the Rashba term [14] (with the coefficient  $\alpha$ ), and of a 2D analogue of the Dresselhaus term (with the coefficient  $\beta$ ) [10,11]. We use the following notations:  $\mathbf{k} = -i\nabla + e\mathbf{A}/c$  and  $\bar{\mathbf{k}} = (k_x, -k_y)$  are 2D vectors;  $\hat{\sigma}^{x,y}$  are the Pauli matrices. The  $\beta$  coefficient is determined by the formula [10,11]:

$$\beta = \gamma_{so} \frac{\hbar}{\sqrt{2m^*G}} \cdot \text{Ry}^* \cdot a_0^2/d^2 \equiv \mathcal{B} \cdot \text{Ry}^* \cdot a_0^3/d^2 , \quad (2)$$

where  $G$  is the band-gap width of the semiconductor,  $\gamma_{so}$  is the spin-orbit constant

[15],  $a_0 = \hbar^2 \varepsilon / m^* e^2$  is the effective Bohr radius. (For GaAs:  $G = 1.52$  eV,  $\gamma_{\text{so}} = 0.07$ ,  $a_0 = 9.95$  nm Ry\*  $\approx 5.74$  meV,  $m^* = 0.067 m_e$ , and  $\mathcal{B} = 0.0043$ .) The parameter  $d$  is determined by averaging the square of the wave-vector  $\hat{z}$ -component of a 2D electron in the layer:  $d^{-2} = -\langle f | d^2/dz^2 | f \rangle$ , where  $f(z)$  is the corresponding size-quantized function. (In the well we consider that  $f = (2/\pi)^{1/4} e^{-z^2/d^2} / \sqrt{d}$ .)

The Hamiltonian of the system of two interacting electrons in the harmonic potential  $\frac{1}{2} m^* \omega_0^2 (x^2 + y^2)$  may be written in Center-of-Mass (CM) and relative coordinates as  $\mathcal{H} = H_m(\mathbf{r}) + \mathcal{H}_M(\mathbf{R}) + \mathcal{H}_S$ . Here  $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$  and  $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$  are Center-of-Mass and relative (Jacobi) coordinates of two particles. The first term in the Hamiltonian is

$$H_m = -\frac{\hbar^2}{2\mu} \left( \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} - \frac{m^2}{r^2} \right) - \frac{\hbar}{2} \omega_c m + \frac{\mu r^2 \omega_0^2}{2} \left( 1 + \frac{h^2}{4} \right) + e^2 / \varepsilon r \quad (3)$$

(here  $\omega_c = eB/m^* c$ ;  $\mu = m^*/2$  is the reduced mass,  $h = \omega_c / \omega_0$  is the dimensionless magnetic field). The expression for  $\mathcal{H}_M(\mathbf{R})$  may be found from Eq. (3) for  $H_m$  after the substitution:  $\mathbf{r} \rightarrow \mathbf{R}$ ,  $\mu \rightarrow \mathcal{M} = 2m^*$ ,  $m \rightarrow M$ , and  $\varepsilon \rightarrow \infty$ .

The term  $\mathcal{H}_S$  is the spin-dependent part of the Hamiltonian, namely it is a combination of the Zeeman and spin-orbit coupling terms:  $\mathcal{H}_S = \sum_{i=1,2} [g\mu_B B |\hat{\sigma}_i^z + H_{SO}^{(i)}]$ . For two electrons, the SO part  $H_{SO}^{(1)} + H_{SO}^{(2)}$  can also be written in CM and relative coordinates  $\mathbf{R}$  and  $\mathbf{r}$ . Denoting  $\hat{\Sigma} = \hat{\sigma}_1 + \hat{\sigma}_2$ ,  $\hat{\sigma} = \hat{\sigma}_1 - \hat{\sigma}_2$ , we obtain

$$\mathcal{H}_S = (H_{RSO} + H_{rSO}) + g\mu_B B \hat{\Sigma}_z, \quad (4)$$

where

$$H_{RSO} = -D_+ (\alpha \hat{\Sigma}_- + i\beta \hat{\Sigma}_+) - D_- (\alpha \hat{\Sigma}_+ - i\beta \hat{\Sigma}_-),$$

and

$$H_{rSO} = -\partial_+ (\alpha \hat{\sigma}_- + i\beta \hat{\sigma}_+) - \partial_- (\alpha \hat{\sigma}_+ - i\beta \hat{\sigma}_-). \quad (5)$$

The new operators are

$$D_{\pm} = \mp \frac{1}{2} \left( \frac{\partial}{\partial X} \pm i \frac{\partial}{\partial Y} \right) + b(X \pm iY), \quad \partial_{\pm} = \mp \left( \frac{\partial}{\partial x} \pm i \frac{\partial}{\partial y} \right) + \frac{b}{2}(x \pm iy)$$

[( $X, Y$ ) and ( $x, y$ ) are the components of 2D vectors  $\mathbf{R}$  and  $\mathbf{r}$ ],  $b = m^* \omega_c / 2\hbar$  and

$$\hat{\Sigma}_{\pm} = \frac{1}{2} (\hat{\Sigma}_x \pm i \hat{\Sigma}_y), \quad \hat{\sigma}_{\pm} = \frac{1}{2} (\hat{\sigma}_x \pm i \hat{\sigma}_y).$$

The wavefunction of the two-electron system may be written in the form  $\Psi_M(R)e^{iM\Phi} \cdot \psi_m(r)e^{im\phi}\chi_{\sigma_1,\sigma_2}$ , where  $\sigma_1$  and  $\sigma_2$  are the spin variables of the electrons. We expand the wavefunction in the basis set of the singlet and triplet states

$$|s\rangle = \Psi_0(R)\psi_0(r)|0,0\rangle \quad \text{and} \quad |t, S_z\rangle = \Psi_0(R)\psi_1(r)e^{i\phi}|1, S_z\rangle \quad (S_z = 0, \pm 1). \quad (6)$$

Here  $\Psi_0$  is the ground state function (i.e. it obeys the equation  $\mathcal{H}_0\Psi_0 = \hbar\omega_0\sqrt{1+h^2/4}\Psi_0$ ), while the functions  $\psi_{0,1}$  have to be found from the equations

$$H_m\psi_m = E_m\psi_m. \quad (7)$$

The analytical solution of Eqs. (7) could be found if  $l = \sqrt{\hbar/m^*\omega_0}(h^2 + 4)^{-1/4} \ll a_0$  or  $l \gg a_0$ . The  $l \ll a_0$  case has been studied perturbatively in Ref. [8]. Here we consider the opposite limit  $l \gg a_0$  (this seems to be more relevant to a typical experimental situation). Then in the leading approximation the solutions of Eqs. (7) are the states of one-dimensional oscillator with mass  $\mu$  and frequency  $\omega_0\sqrt{3+3h^2/4}$  localized in the vicinity of  $r_0 = [2l_0^4/a_0(1+h^2/4)]^{1/3} \gg l_0$  (here we have designated  $l_0 = \sqrt{\hbar/m^*\omega_0}$ ). In this approximation the energy measured from the ground state is

$$\delta_{m,S_z} = E_m - E_0 = \frac{\hbar^2 m^2}{r_0^2 m^*} - \frac{\hbar}{2}\omega_c(m + g^*S_z) \equiv \frac{\hbar\omega_0}{4}D_{m,S_z}(h) \quad (8)$$

( $g^* = gm^*/m_e \approx 0.029$ ). The equation determining the ST crossing takes thereby the form  $\delta_{1,S_z} = 0$  (if  $a_0/l_0 = 1/3$ , then for the ST crossing point we obtain  $h = 0.64$  at  $S_z = 0$ , whereas for the same  $a_0/l_0$  and  $S_z$  the exact numerical calculation [8] gives  $h = 0.69$ ).

We now turn to the effect of SO interaction on the mixing of singlet and triplet states. Operators  $\Sigma_{\pm}$  and  $\Sigma_z$  commute with  $\Sigma^2$ , therefore the first and third terms in Eq. (4) are not responsible for mixing of the singlet and triplet states. On the contrary, the second term in Eq. (4) results in this mixing. Indeed, let  $|S, S_z\rangle$  be the normalized spin states of two electrons. When operating on the state  $|0,0\rangle$ , the term  $H_{rSO}$  yields the following non-zero matrix elements:  $\langle 1, 1|H_{rSO}|0,0\rangle$  and  $\langle 1, -1|H_{rSO}|0,0\rangle$ . (One can check that  $\hat{\sigma}_{\pm}|0,0\rangle = \mp 2^{-1/2}|1, \pm 1\rangle$ .)

Hence we see immediately the  $|0,0\rangle$  singlet state is mixed with the  $|1, \pm 1\rangle$  triplet states but not with the  $|1,0\rangle$  state. This state is therefore long-lived.

For the states which are coupled, the expansion in terms of the small parameter  $a_0/l_0$  leads to the following result for the mixing matrix element between  $|s\rangle$  and  $|t, 1\rangle$  states:

$$M_{SO} = \langle s|H_{rSO}|t, 1\rangle \approx -\frac{i\beta}{2^{3/2}} \left( \frac{1}{r_0} + br_0 \right) \equiv -\frac{i\beta}{2^{3/2}l_0} \mathcal{L}(h). \quad (9)$$

If we take into account the Rashba term, we would find that another non-zero matrix element is  $\langle s|H_{rSO}|t, -1\rangle$ , however *the state  $|t, 0\rangle$  is never mixed with the singlet*. Further we neglect the Rashba coupling (usually there is  $\alpha < \beta$  in GaAs hetero-structures, besides  $\alpha$  vanishes in the case of an ideally symmetric quantum well).

With help of Eq. (9) we find the hybridized states  $|S\rangle = C_0^-|s\rangle + C_1^-|t, 1\rangle$ , and  $|T\rangle = C_0^+|s\rangle + C_1^+|t, 1\rangle$ , where

$$C_0^\pm = \left\{ \frac{M_{SO}}{2|M_{SO}|} \left[ 1 \mp \frac{\delta_{1,1}}{\delta E} \right] \right\}^{1/2}, \quad C_1^\pm = \pm \left\{ \frac{M_{SO}}{2|M_{SO}|} \left[ 1 \pm \frac{\delta_{1,1}}{\delta E} \right] \right\}^{1/2} \quad (10)$$

$(\delta E = \sqrt{\delta_{1,1}^2 + 4|M_{SO}|^2})$ . The corresponding energies are  $E_{T/S} = E_0 + (\delta_{1,1} \pm \delta E)/2$ .

The next step in our study is the calculation of the  $|T\rangle \rightarrow |S\rangle$  (or  $|S\rangle \rightarrow |T\rangle$ ) relaxation rate for the case, where  $\delta_{1,1} > 0$  (or  $\delta_{1,1} < 0$ ). Evidently the main relaxation channel is determined by emission of the acoustic phonon with energy  $\hbar c_s k = \delta E$ ; where  $\mathbf{k} = (\mathbf{q}, k_z)$  is the phonon wave vector, and  $c_s$  is the mean sound velocity (we use the so-called isotropic model, i.e.,  $c_s$  does not depend on the polarization and on the  $\mathbf{k}$  direction; we consider that  $c_s = 3.37 \cdot 10^5$  cm/s). The probability of this event is determined by

$$\frac{1}{\tau} = \sum_{\mathbf{q}, k_z} \frac{2\pi |\mathcal{M}(\mathbf{q}, k_z)|^2}{\hbar} \delta(\hbar c_s k - \delta E), \quad (11)$$

where  $\mathcal{M}(\mathbf{q}, k_z)$  is the appropriate matrix element

$$\mathcal{M}(\mathbf{q}, k_z) = \langle T|U_{\text{e-ph}}|S\rangle = C_0^{+*} C_0^- (\langle s|U_{\text{e-ph}}|s\rangle - \langle 1, t|U_{\text{e-ph}}|t, 1\rangle). \quad (12)$$

Here the phonon field is averaged over the angle  $\phi = r \wedge q$ :

$$U_{\text{e-ph}}(\mathbf{R}, r) = \left( \frac{\hbar}{V} \right)^{1/2} \sum_s \tilde{U}_s(\mathbf{q}, k_z) e^{i\mathbf{q}\mathbf{R}} \overline{(e^{i\mathbf{q}\mathbf{r}} + e^{-i\mathbf{q}\mathbf{r}})} = 2 \left( \frac{\hbar}{V} \right)^{1/2} \sum_s \tilde{U}_s(\mathbf{q}, k_z) e^{i\mathbf{q}\mathbf{R}} J_0(qr) \quad (13)$$

[ $s$  is the polarization,  $V$  is the sample volume, and  $\tilde{U}_s(\mathbf{q}, k_z)$  is the renormalized vertex which includes the deformation and piezoelectric fields created by the phonon]. The integration

with respect to  $z$  has already been performed, and reduces to the renormalization  $\tilde{U}_s = U_s(\mathbf{q}, k_z)F(k_z)$ , where the formfactor is  $F(k_z) = \langle f | e^{ik_z z} | f \rangle$ . By using Eqs. (6), (12) and the expansion  $J_0(qr) \approx J(qr_0) - q(r - r_0)J_1(qr_0)$ , we obtain the matrix element (12); and after the substitution into Eq. (11) we find that the relaxation rate is proportional to the  $|\sum_s U_s|^2$ . The latter is represented as [12]

$$\left| \sum_s U_s(\mathbf{q}, k_z) \right|^2 = \frac{\pi \hbar c_s k}{p_0^3 \tau_A}, \quad \text{where} \quad \frac{1}{\tau_A(\mathbf{k})} = \frac{1}{\tau_D} + \frac{5p_0^2}{k^6 \tau_P} (q^2 k_z^2 + q_x^2 q_y^2). \quad (14)$$

The summation involves averaging over the directions of the polarization unit vectors for both components of the electron-phonon interaction. The nominal times for the deformation and piezoelectric interactions in GaAs are  $\tau_D \approx 0.8$  ps, and  $\tau_P \approx 35$  ps [12,16]. The nominal momentum is  $p_0 = 2.52 \cdot 10^6$ /cm [12,16]. We also refer to Refs. [12,16] for details concerning the meaning and the expressions of these values in terms of the GaAs material parameters.

Finally, with the help of Eqs. (2),(9),(10),(12)-(14) and (11) we calculate the relaxation time.

$$1/\tau(h) = \mathcal{W} \cdot \mathcal{E} \mathcal{L}^2 \cdot \exp(-\mathcal{A} \mathcal{E}^2) \cdot \mathcal{J}.$$

where

$$\mathcal{W} = \frac{\pi(\text{Ry})^3 a_0^{10} \mathcal{B}^2}{216(\hbar c_s)^3 p_0 \tau_P l_0^4 d^4}, \quad \mathcal{E}(h) = \frac{4\delta}{\hbar \omega_0} = \sqrt{\mathcal{D}_{1,1}^2(h) + 2[\mathcal{B} a_0 l_0 \mathcal{L}(h)/d^2]^2}$$

[see the definitions for  $\mathcal{L}$  and  $\mathcal{D}_{1,1}$  in Eqs. (8-9)],

$$\mathcal{A} = \left( \frac{d a_0^2 \text{Ry}}{4 l_0^2 \hbar c_s} \right)^2, \quad \mathcal{J}(h) = \int_0^1 \frac{d\xi \xi}{\sqrt{1-\xi}} \{J_1[\mathcal{R}(\xi)]\}^2 \left[ 5\xi - \frac{35}{8}\xi^2 + \mathcal{S} \mathcal{E}^2(h) \right] e^{-\xi \mathcal{P}(h)},$$

$$\mathcal{R}(\xi) = a_0 \text{Ry} / \hbar c_s (a_0 / l_0)^{2/3} \sqrt{\xi \mathcal{E}(h) (4 + h^2)^{-1/3}}, \quad \mathcal{S} = (\tau_P / \tau_D) \left( a_0^2 \text{Ry} / 2 l_0^2 \hbar c_s p_0 \right)^2,$$

and

$$\mathcal{P}(h) = \left[ a_0^2 / l_0^2 \sqrt{4 + h^2} - \frac{1}{2} \left( d a_0 / l_0^2 \right)^2 \right] [\mathcal{E}(h) a_0 \text{Ry}]^2 / 8 (\hbar c_s)^2.$$

As an illustration Fig. 1 shows the relaxation rate as a function of the magnetic field on the logarithmic scale (the main picture) and in the usual scale (the inset). The relaxation time is seen to have a sharp maximum in the vicinity of the ST crossing but constitutes a comparatively small value (of the order of 0.1 mcs) in the regions where the singlet and triplet lines are resolved. The non-monotonic behaviour of  $\tau$  on the right of the ST crossing

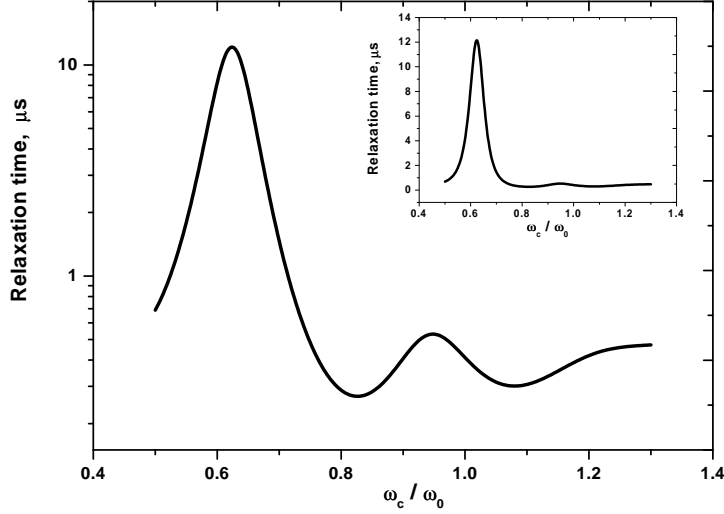


FIG. 1. The ST relaxation time  $\tau$  calculated for  $a_0/l_0 = 1/3$  and  $d = 5$  nm. The maximum corresponds to ST transition at  $\omega_c/\omega_0 = 0.64$ . See the text for details.

originates from the correlations between the wave-function characteristic distance  $r_0$  and the wavelength  $\hbar c_s/\delta E$  of the emitted phonon.

We now turn to the discussion of the manner in which the ST relaxation could influence the transport spectroscopy through the QD states. By studying the kinetic processes of filling and emptying the dot in the presence of a large “source-drain” voltage, we estimate the effective electron life-time inside the dot at the “working level” i.e. at the level which participates in the transport through the dot. This effective life-time  $\tau_{\text{dot}}$  under experimental conditions of Refs. [6,7] we estimate to be of the order of 1 mcs, and this value should evidently be compared with the ST relaxation time calculated above. If the working level is exactly the upper level of the two-electron droplet, then the relaxation could influence the current. Namely, if  $\tau < \tau_{\text{dot}}$ , then the working level could be emptied due to the ST relaxation occurring within the dot. In this case the current through the upper two-electron state becomes negligible. The relaxation process is asymmetric across the ST transition. Before the transition the “working level” involves  $|t, \pm 1\rangle$  and  $|t, 0\rangle$  [see Eq. (6)] triplet states. (The Zeeman splitting is not resolved.) The  $|t, 0\rangle$  state is long lived and hence observed in experiment while the  $|t, 1\rangle$  state relaxes efficiently to the  $S = 0$  singlet state

$|s\rangle$ . Past the ST transition, the  $|t, 1\rangle$  state is the ground state but the excited state is the singlet. The singlet state relaxes efficiently to the  $|t, 1\rangle$  ground state. Hence this asymmetry in the relaxation processes associated with the singlet-triplet transition could be responsible for the anomalies observed in transport experiments [6,7].

In closing it is worthy to mention other relaxation channels which are not taken into account in our calculation but which in the framework of the considered mechanism could additionally reduce the ST relaxation time. These are provided by special phonon modes (e.g., by surface and confinement phonons excited in the hetero-junction) and certainly by the SO Rashba coupling if the latter is significant.

The authors acknowledge A.Sachrajda, D.G. Austing and J. Kyriakidis for discussions. S.D. acknowledges the support of the Russian Foundation for Basic Research and thanks the Institute for Microstructural Science (Ottawa), where this project was initiated, for hospitality.

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